

Crustal controls on apparent mantle pyroxenite signals in ocean-island basalts

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ABSTRACT

Ocean-island basalts (OIBs) provide a unique insight into the extent of lithological heterogeneity (peridotite vs pyroxenite) in the Earth's convecting mantle. However, crustal processing of these mantle melts significantly influences minor-element concentrations in olivine phenocrysts, challenging the suitability of this widely-used approach to identify lithological variations in their mantle source. Using a numerical model of magma recharge, mixing and diffusional re-equilibration we show that this type of crustal processing -- which is widely observed in OIBs -- results in elevated Ni and lower Ca contents of Fo-rich olivine causing erroneously high estimates of the proportion of pyroxenite-derived melt.

We applied our model of magma recharge and mixing to several OIBs including the Galápagos, Canaries and La Réunion. In particular, we critically examine olivine compositional variations in basalts from the eastern Galápagos, which display Sr- and Pb-isotope ratios similar to N-MORBs. Whilst previous interpretations (based on olivine chemistry) argue for a significant contribution from pyroxenite-derived melt, our results indicate that the postulated presence of pyroxenite in the eastern Galápagos mantle is an artefact of processing of magmas and their olivine cargo as they transition through the crust, consistent with major element and isotopic evidence for a dominantly peridotitic source in this region.

This new model for magma recharge and mixing may have important implications for our understanding of lithological heterogeneity beneath OIBs globally, and highlights the importance of considering crustal processes when attempting to interpret olivine compositions with regard to mantle heterogeneity.

INTRODUCTION

Entrainment and subsequent melting of recycled oceanic lithosphere in upwelling mantle plumes has long been recognized as an important process resulting in the varied array of isotopic signatures displayed by ocean island basalts (OIBs; Hofmann and White 1982; Stracke et al. 2005). During plume upwelling entrained oceanic crust (eclogite) will melt at depths greater than the peridotite solidus and these melts may react with the host peridotite to form pyroxenite (Yaxley and Green, 1998). The extent of this lithological variation in the mantle source regions of OIBs is widely debated (Sobolev et al., 2005; Herzberg et al., 2016; Matzen et al., 2013; 2017a,b) but is of critical importance as the amount of pyroxenite influences the thermodynamic properties of upwelling mantle, as well as the chemical and rheological characteristics of melts formed during adiabatic decompression (Herzberg, 2011).

While the presence of pyroxenite in the mantle source of OIBs has been identified based on major element compositions (Hauri, 1996), and variations in isotopic ratios (Day et al., 2009), the most common method involves analysis of minor element (Ni, Mn, Ca) concentrations in olivine phenocrysts (Sobolev et al. 2007; Herzberg et al., 2014). The concentration of Ni, Mn and Ca in primary mantle melts is controlled by their bulk partition coefficients during mantle melting. Whilst it has been shown that temperature and melt composition influence the partitioning of Ni and Mn between olivine and melt (Putirka et al., 2011; Matzen et al., 2017a,b), the relative proportions of olivine and pyroxene in the mantle source plays a critical role in

determining the bulk partition coefficients for these elements (Sobolev et al., 2007). As a result, it has been suggested that high Ni and low Mn and Ca contents observed in Hawaiian olivines result from melting of a pyroxene-rich mantle source (i.e. pyroxenite; Sobolev et al., 2007; Herzberg, 2011), whereas lower Ni contents in olivines from MORBs are consistent with low- P melting of an olivine-rich lithology (i.e. peridotite; Fig. 1; Sobolev et al., 2007).

Recent studies have, however, concluded that post-melt generation processes may influence the minor-element chemistry of olivine in OIBs. For example, Hole (2018) showed that high- P crystallization of primary mantle melts may result in olivine Ni contents that are elevated above the typical crystal-line-of-descent curves estimated for peridotite-derived melts, due to the influence of augite fractionation. Deep fractionation of augite may also influence the Mn, and therefore the Fe/Mn ratio, of basaltic magmas and their derivative olivines (e.g. Vidito et al., 2013). However, this effect is likely to be significantly smaller than for Ni and Ca once uncertainty in Fe/Mn measurements are considered. Hole (2018) suggests that crystallization at >1.6 GPa may explain the high Ni and low Ca contents of olivines in some flood basalt provinces. Since this crystallization pressure is higher than normally encountered in OIBs we explore other post-melt generation processes that might impact the minor element concentrations of olivine.

NUMERICAL MODEL OF MAGMA RECHARGE AND MIXING

Mafic magma recharge and mixing is a common process in many OIBs (e.g. Neumann et al. 1999). As a result, we have developed a simple numerical model to investigate the influence of these processes on the minor element concentrations in olivine. Our models are based on a series of crystallization models in Petrolog (Danyshevsky and Plechov, 2011) and combine fractional crystallisation with magma mixing and diffusive re-equilibration. We use

parameterisations for partitioning of: (i) Ni and Ca into olivine from Beattie et al. (1991) and Herzberg and O'Hara (2002), respectively; and (ii) Ni into clinopyroxene from Mysen (1978). Crystallisation models are run at pressures appropriate for each ocean island considered (Geist et al., 1998; Albarede et al., 1997).

Primitive (high Mg#) basalts from each ocean island are fractionation corrected to be in equilibrium with mantle peridotite and set as the starting composition. The initial Ni content of the melt is calculated using the method of Matzen et al. (2017a; Supplementary File). To simulate magma recharge events a primitive basaltic melt (chemically equivalent to the initial melt present in the system) is introduced into the system at set temperature intervals and allowed to mix with the fractionated magma (Fig. 2). The temperature interval between each period of recharge (i.e. the extent of cooling) was assumed to be greater during olivine-only crystallization than during fractionation of a gabbroic assemblage. This is due to the smaller change in temperature with respect to changes in enthalpy of the magma (i.e. heat loss) during gabbroic fractionation (Ghiorso, 1997). After mixing, the hybrid magma continues to cool and crystallise until the next injection of primitive mafic magma occurs. The mass of primitive melt added during each recharge event is assumed to be equivalent to the initial mass of magma present in the system.

We also investigate the influence that these periods of recharge may have on the composition of previously formed olivine crystals present in the system. For example, mafic recharge may result in disequilibrium between olivines and the new melt compositions, resulting in resorption of these crystals prior to further crystallization. In addition, diffusion may rapidly modify the initial composition of these crystals. By developing a simple numerical model of chemical diffusion in olivine (using coefficients from Chakraborty, 2010), we place constraints

on the timescales required for the olivine compositions to diffusively re-equilibrate with the hybrid magma composition following magma recharge. Our model simulates the re-equilibration of a population of high-Fo olivines, formed during the initial stages of crystallisation of a peridotite-derived melt, with a magma formed via mixing and homogenisation of a primitive mantle melt and a more evolved basaltic melt (Fig. 1b). Numerical solutions to the diffusion equation are used to simulate re-equilibration of olivine phenocrysts with the hybrid melt for 1, 10 and 30 years. At each time interval the model olivines were randomly sectioned and their apparent core compositions were recorded; this was done to simulate the influence of random sectioning of natural olivine crystals. The results show that almost complete re-equilibration of Fo, Ni and Mn with the hybrid melt composition may occur in as little as 30 years for 1mm sized olivines, with slightly longer timescales required for slower diffusing elements such as Ca.

IMPLICATIONS OF MAGMA RECHARGE AND MIXING

Whilst magma mixing and recharge cannot explain the high (>4000 ppm) Ni contents of some high Fo (~90) Hawaiian olivines, a first-order observation of our model is that the Ni and Ca contents of olivine phenocrysts with Fo<88 can be significantly influenced by crustal processing of basaltic magmas as they transition through the crust. Olivines crystallising from mixed magmas are predicted to have elevated Ni and low Ca contents resulting in an apparent ‘pyroxenite signal’. In addition, the results of our diffusion modelling show that any previously formed olivines would also acquire this signature over relatively short timescales. Below we explore the implications of these observations for several OIBs.

Eastern Galápagos Basalts

Basalts erupted in the eastern Galápagos (i.e. Isla Santa Cruz and eastern Santiago) are typically derived from an isotopically-depleted mantle source, similar to that sampled at mid-

ocean-ridges (Harpp and White, 2001; Hoernle et al., 2000; Gibson et al., 2012; Fig. S.1). However, previous interpretations of olivine chemistry suggest that there is a significant contribution of pyroxenite-derived melts to these basalts (Vidito et al., 2013). These interpretations contradict major-element concentrations and isotope ratios consistent with a peridotite source (Supplementary File; Gibson et al., 2012). To investigate this apparent paradox, we present high-precision analyses of olivine phenocrysts hosted in basalts from the eastern Galápagos. These analyses are interpreted alongside our model of magma recharge and mixing. To evaluate the possible presence of pyroxenite in the source region of these basalts we consider the parameters Fe/Mn and Ni*FeO/MgO, which have been proposed to be sensitive to variations in source lithology, but insensitive to the effects of fractional crystallization (Gurenko et al., 2009).

Fe/Mn ratios (64.8 ± 6.3) of olivines from these eastern Galápagos basalts are consistent with those expected to crystallise from peridotite-derived melts (Fig. 3; Herzberg, 2011). However, whilst the Fe/Mn ratios of each sample are equivalent within analytical error, significant differences in Ni*FeO/MgO are seen. At first sight, these results suggest that some eastern Galápagos basalts (07DSG82 & 07DSG83; Fig. 3) contain a significant proportion of pyroxenite-derived melt. However, we show that the intra-sample variability may instead be caused by variations in the crustal processing of these magmas.

The compositions of olivines in individual flows follow parallel trends in Fo vs Ni space, which are consistent with fractional crystallization. Most samples display similar maximum and minimum Ni contents but are offset by their Fo contents, such that at ~2000 ppm Ni the observed Fo content of olivines in adjacent flows from Isla Santa Cruz vary from ~84 to ~86.5. Crucially, our numerical models of magma recharge predict a similar relationship, i.e. that the range in

olivine Ni contents following each period of recharge is constant, and only changes in the Fo content are observed. Consequently, the $\text{Ni}^*\text{FeO}/\text{MgO}$ content of olivine increases following each period of recharge.

The olivine compositions predicted by our magma recharge models for peridotite-derived melts accurately match the composition of olivine populations observed in isotopically-depleted basalts from the eastern Galápagos (Fig. 3). Additionally, we show that the compositions of any olivines formed from primary magmas are rapidly overprinted by diffusive processes during storage and/or transport of these olivines following magma hybridisation. Our numerical models of magma recharge and mixing are in keeping with the presence of the large ($>2\text{mm}$), reversely-zoned olivine crystals in these basalts (Fig. S.3). These crystals record lower core $\text{Ni}^*\text{FeO}/\text{MgO}$ values than their high-Fo mantles; for example, olivine #3 from sample 07DSG83 shows core compositions with $\text{Ni}^*\text{FeO}/\text{MgO}$ values of ~ 400 , and high-Fo mantles with $\text{Ni}^*\text{FeO}/\text{MgO}$ values of ~ 550 (orange line in Fig. 3). This variation is consistent with diffusive re-equilibration (over several decades) following magma mixing and provides compelling evidence that mixing and recharge of melts from a peridotite source is responsible for the high Ni contents of olivine populations in the eastern Galápagos.

Our findings suggest that there is no need for the isotopically-depleted eastern Galápagos plume component to consist of pyroxenite, consistent with major-element and isotopic evidence (Supplementary information; Gibson et al., 2012). However, we cannot exclude the presence of pyroxenite source lithologies in other (isotopically enriched) regions of the Galápagos (Vidito et al., 2013).

Broader implications

Overestimation of the proportion of pyroxenite-derived melt due to magma recharge is not restricted to the Galápagos, and our results have implications for how source lithology is inferred at other volcanic islands. For example, olivine compositions from Tenerife and La Palma in the Canary Islands (Gurenko et al., 2009) and La Réunion (Sobolev et al., 2007) are successfully reproduced using our model of magma recharge and mixing. The data from La Réunion is best reproduced under high-pressure crystallisation, consistent with previous interpretations (Albarede et al. 1997). Additionally, ^{226}Ra - ^{230}Th disequilibria at Piton de la Fournaise indicate that multiple periods of magma recharge occur, and magma residence times are on the order of decades to centuries (Sigmarsson et al. 2005). Similar observations related to magma recharge have been made for basaltic systems in the Canary Archipelago (Neumann et al., 1999). These results are in keeping with our proposed model and thus indicate that magma mixing may have an important role in controlling the olivine chemistry of lavas from these islands. As a result, previous estimates of ~50% pyroxenite-derived melt contribution to lavas erupted on these islands (Sobolev et al., 2007; Gurenko et al., 2009) should only be considered as a maximum estimate.

Our results reveal that open-system magmatic processes may have a significant effect on the minor-element concentrations of olivine crystallising from mantle melts. Additionally, diffusion models reveal that 1mm olivine crystals undergo almost complete equilibration with the new melt composition within only 30 years (Fig. 1b). This rapid modification of olivine compositions following magma recharge in basaltic systems is significant as it shows that we are unlikely to observe olivine compositions in equilibrium with the primary mantle melt.

The influence of these processes is be greatest on olivine populations from any system where the ratio between magma flux and cooling rate allows relatively small ephemeral magma storage regions to exist on timescales of several centuries (e.g. eastern Galápagos; Saal et al. 2000; Gibson et al., 2016), yet is not so high that large-scale compositionally buffered magmatic systems are maintained over thousands of years. In relatively small magmatic systems, where magma recharge events occur infrequently and may represent a significant fraction of the magmatic system, substantial changes in melt chemistry may occur. As shown in the models above, these changes result in formation of olivine crystals with elevated Ni contents. However, in regions of high melt-flux (e.g. MORBs), where steady-state magma chambers are maintained over a narrow compositional range by eruption, crystallisation and replenishment (O'Neill and Jenner, 2012), similar enrichment in olivine Ni contents are not predicted (Supplementary File). We therefore conclude that crustal processes such as magma recharge must be considered, especially for OIBs, otherwise significant overestimation of the proportion of pyroxenite-derived melt may occur.

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294

FIGURE CAPTIONS

Figure 1 – Global variability in olivine Ni and Fe/Mn contents are shown using data for Koolau, Hawaii and MORBs from Sobolev et al. (2007). Koolau olivines display the high Ni and Fe/Mn contents believed to be diagnostic of pyroxenite-derived melts, whereas MORB olivines display low Ni and Fe/Mn values typical of low-pressure melting of peridotite. It should be noted that high-pressure melting of peridotite can result in higher Ni and Fe/Mn contents in olivines than observed in MORBs. As an example, the red star in **A.** represents a peridotite source solution for melting at ~2.5 GPa using the model of Matzen et al. (2017a). Using this peridotite source solution as a starting point the olivine compositions from La Réunion are shown to be consistent with a peridotite source, once the influence of magma recharge and mixing is accounted for (orange dashed lines). Low-pressure primary magma solutions for increasing proportions of pyroxenite derived melt (X_{PX}) from Vidito et al. (2013) are shown ranging from 0 – 1.

Figure 2 – **A.** Fo vs Ni graph showing the influence of repeated mafic recharge on the Ni content of magmatic olivines. Mixing trajectories are shown by the orange dashed lines. Melt Ni content and temperature at each labelled point are: (1) 1365°C, 515 ppm; (2) 1166°C, 73.4 ppm; (3) 1284°C, 288 ppm; (4) 1137°C, 48.7 ppm; (5) 1261°C, 253 ppm; (6) 1111°C, 41 ppm; (7) 1242°C, 237 ppm; and (8) 1095°C, 39.4 ppm. Temperature intervals are ~200°C when olivine-only fractionation occurs and ~150°C when clinopyroxene and plagioclase become crystallising phases. **B.** Re-equilibration of 1 mm olivine crystals from the initial fractionation curve with a melt composition resulting from magma mixing. Point (5) is chosen to represent the equilibrium melt composition rather than point (3) as points (1) and (3) have very similar compositions, and thus it is not easy to visualize the extent of re-equilibration of these olivines with a melt composition in equilibrium with point (3).

Figure 3 –Ni, Fe/Mn and Ca contents of isotopically-depleted basalts from eastern Galápagos are consistent with fractional crystallisation and mafic recharge of a peridotite-derived primary magma. Model results (black lines) are identical to those shown in Fig. 2. Dashed region in **A.** represents data for eastern Galápagos basalts from Gibson et al. (2016). Bold orange line in panels **A.** and **D.** show the zoning profile displayed by olivine #3 in sample 07DSG83. This profile varies from a core composition of Fo ~ 78, Ni*FeO/MgO ~ 400 to a mantle composition of Fo ~ 84, Ni*FeO/MgO ~ 550.

Figure 1

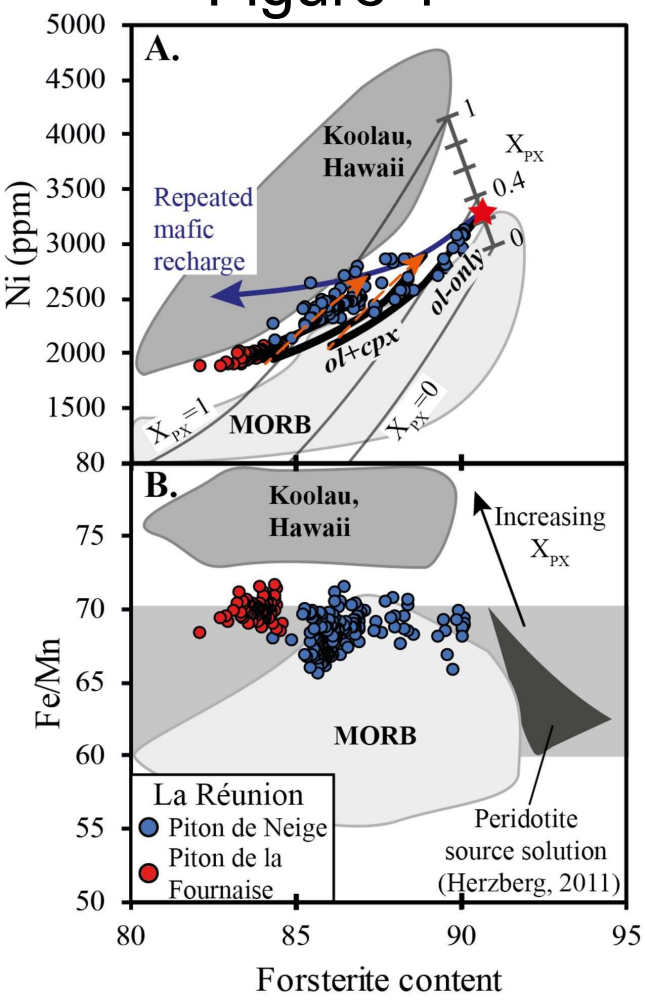


Figure 2

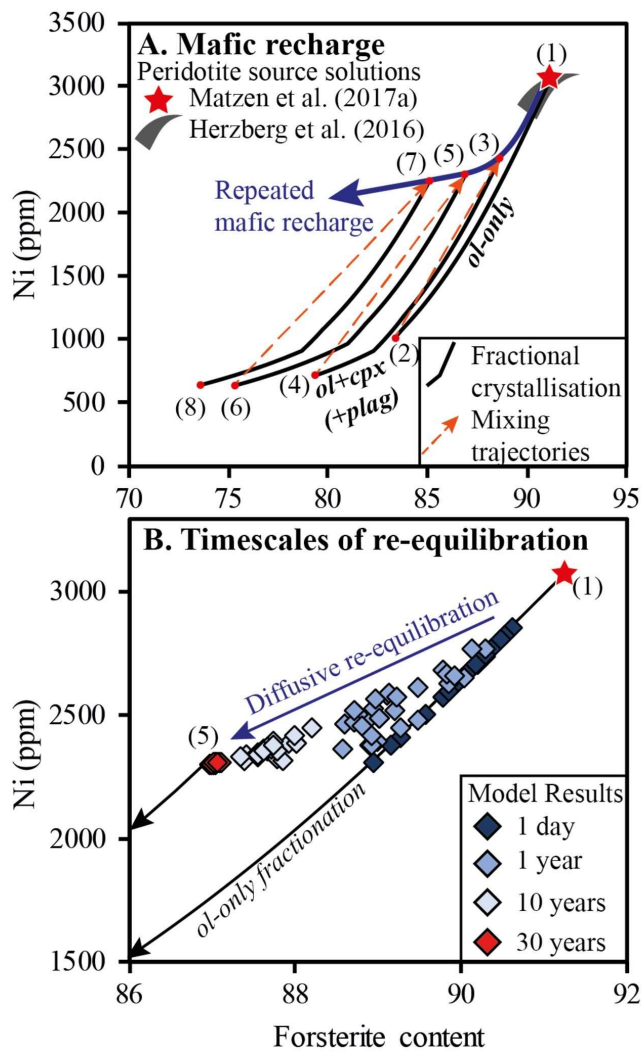


Figure 3

